



# Operational characteristics of efficient co-removal of H<sub>2</sub>S and NH<sub>3</sub> in a horizontal biotrickling filter using exhausted carbon

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## ABSTRACT

Odorless H<sub>2</sub>S and NH<sub>3</sub> gases were effectively biodegraded in a horizontal biotrickling filter (HBTF) packed with H<sub>2</sub>S-exhausted carbon. During the process, the operational characteristics including the distribution of degradation products, biomass accumulation and biological activity, carbon surface characteristics and pressure drop were investigated. The results show that the content of biodegradation products on carbon was low, i.e. 0.9–2.8 wt% S and 0.3–1.0 wt% N. The low content benefited the stable operation of the HBTF, due to preventing a toxic concentration of degradation products on packing bed over a long-term operation. The biomass was distributed evenly along the HBTF. This avoided the problems of bed clogging and activity loss. On the other hand, the deteriorated performance was observed due to the biomass accumulation over a long-term operation. Carbon surface characteristics in the HBTF remained almost unchanged. pH values of carbon were neutral and micropore structure of carbon remained relatively stable. In addition, the pressure drop in the HBTF was very low. These operational characteristics of the HBTF system significantly contributed to efficient co-removal of H<sub>2</sub>S and NH<sub>3</sub> in the HBTF over a long-term operation.

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## 1. Introduction

Biological treatment is used increasingly as a method for treating large flow rates and low concentrations of waste gases streams containing odors or volatile organic compounds (VOCs), because it is cost-effective and environmental friendly compared with the conventional physical and chemical techniques [1]. The two most promising bioreactors for the control of waste gases are biofilters and biotrickling filters. Biofilters have been used successfully for odor abatement in the last few decades, while waste air treatment using biotrickling filters is a relatively new technique. In biofilters, the contaminated air passes through a moist bed generally packed with natural material. In biotrickling filters, the polluted air, together with a re-circulated liquid, passes through a packed bed on which a pollutant-degrading biofilm develops. The presence of trickling liquid phase in biotrickling filters allows better control of operation conditions [2].

H<sub>2</sub>S and NH<sub>3</sub> are the major contributors of sewage odor with very low odor thresholds (1.1 ppb<sub>v</sub> for H<sub>2</sub>S and 37 ppb<sub>v</sub> for NH<sub>3</sub>) and high toxicity [3]. Biofilters have been widely used for the co-removal of H<sub>2</sub>S and NH<sub>3</sub> [3–6]. A long empty bed residence time (EBRT), e.g., 20–180 s, was usually required to achieve high removal

efficiency. Biodegradation capacity of NH<sub>3</sub> was significantly suppressed by high loadings of H<sub>2</sub>S or NH<sub>3</sub>. The poor performance could be attributed to substantial operational problems existed in biofilters. Firstly, the accumulation of degradation products on packing materials may inhibit the biodegradation capacity [3,7,8]. Secondly, biofilters often get acidified due to the production of sulfate and/or nitrite/nitrate by the oxidation of H<sub>2</sub>S and/or NH<sub>3</sub>. This in turn inhibits the biofilm activity [5,9]. Moreover, biofilters bed was found to dry out easily [10]. In addition, the removal of H<sub>2</sub>S and NH<sub>3</sub> might be affected by biomass accumulation and activity loss in biofilters, even though little attention has been given to this issue [11].

In a biotrickling filter, the recirculation liquid could remove metabolic products on the packing, and the packing acidification can be avoided due to better control of pH [12]. Drying out of the packing bed is not an issue in biotrickling filters. Moreover, biotrickling filter is an attractive technology for the removal of NH<sub>3</sub> because of its high water solubility. Recently, biotrickling filters have been used for the co-removal of H<sub>2</sub>S and NH<sub>3</sub> [13–15]. The biodegradation capacity of NH<sub>3</sub> and H<sub>2</sub>S was improved in the biotrickling filters, especially with optimal substrates acclimation strategy and using H<sub>2</sub>S-exhausted carbon as packing material [14,16]. Almost complete removal of NH<sub>3</sub> and H<sub>2</sub>S was achieved at EBRT longer than 4 and 8 s, respectively. Moreover, high removal of NH<sub>3</sub> was obtained at high loadings of H<sub>2</sub>S and NH<sub>3</sub> up to 137 g m<sup>-3</sup> h<sup>-1</sup> [14].

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However, these studies mostly focused on evaluating removal efficiency, removal capacity, and removal kinetics, while few studies have been found on the operational characteristics in a biotrickling filter for the co-removal of  $\text{H}_2\text{S}$  and  $\text{NH}_3$ . Understanding system operational characteristics in bioreactors is highly relevant for improving removal performance of contaminants from gas streams. Moreover, understanding these issues allow for proper design and optimization of operational parameters responsible for the effectiveness of biological odor treatment in a biotrickling filter.

Thus, the present study focuses on evaluating the operational characteristics in a horizontal biotrickling filter (HBTF) packed with exhausted carbon. The general performance of the HBTF for the effective removal of  $\text{H}_2\text{S}$  and  $\text{NH}_3$  has been described previously [14]. Here, the operational parameters during the process including the distribution of degradation products, biomass accumulation and biological activity, carbon surface characteristics and pressure drop were investigated.

## 2. Materials and methods

### 2.1. Microbial immobilization

Activated sludge stream at the secondary sedimentation tank in a local wastewater treatment plant was used for microbial enrichment. The detailed procedure for enriching sulfide-oxidizing bacteria (SOB) consortium was described previously [10]. Nitrifying microbial consortium was enriched by refreshing the medium every day for 30 days, and the medium consisted of ( $\text{g L}^{-1}$ ):  $\text{NH}_4\text{Cl-N}$ , 0.2;  $\text{KH}_2\text{PO}_4$ , 0.2;  $\text{K}_2\text{HPO}_4$ , 0.1;  $\text{NaHCO}_3$ , 2.0; and 10 mL of trace element solution in 1 L of medium. The element solution consisted of ( $\text{g L}^{-1}$ ):  $\text{Na}_2\text{-EDTA}$ , 1.0;  $\text{FeCl}_2\cdot 4\text{H}_2\text{O}$ , 1.0;  $\text{CuCl}_2\cdot 2\text{H}_2\text{O}$ , 0.1;  $\text{ZnSO}_4\cdot 7\text{H}_2\text{O}$ , 1.1;  $\text{MnCl}_2\cdot 4\text{H}_2\text{O}$ , 0.3;  $\text{CoCl}_2\cdot 7\text{H}_2\text{O}$ , 0.1;  $\text{MgCl}_2\cdot 6\text{H}_2\text{O}$ , 2.0;  $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$ , 0.1;  $\text{CaCl}_2\cdot 2\text{H}_2\text{O}$ , 2.5; and  $\text{MgCl}_2\cdot 6\text{H}_2\text{O}$ , 0.2.

The  $\text{H}_2\text{S}$ -exhausted carbon (AddSorb VA3, pellet-shaped in 4 mm of diameter, Jacobi Group) was obtained as described previously [17]. Activated carbon was packed in a glass tube (4.8 cm

i.d.) at a height of 22.9 cm. 1% (v/v)  $\text{H}_2\text{S}$  together with moist air (RH 80%) was diverted into the bottom of the carbon bed. The empty bed retention time was 4.8 s. The experiments were carried out at room temperature. The outlet  $\text{H}_2\text{S}$  concentration was monitored until a breakthrough at 50 ppmv. The main properties of the exhausted carbon were as follows: S content (w/w, %), 7.8; pH value, 6.15; surface area ( $\text{m}^2\text{g}^{-1}$ ), 776; bulk density ( $\text{kg m}^{-3}$ ), 772. 1.7 kg of exhausted carbon was stuffed into the HBTF randomly, and 200 mL of enriched nitrifying consortium ( $2400\text{ mg L}^{-1}$ ) was poured into from the top of the HBTF. This inoculation was performed by continuously trickling the ammonium growth medium over the bed. The nitrifying culture was grown and attached on the carbon surface. After 20 days, the reactor showed a good ability of biodegrading ammonium in the liquid medium and a thin layer of biofilm were observed (not shown here). Separately, 1.7 kg of exhausted carbon was soaked in 2 L of SOB consortium ( $1100\text{ mg L}^{-1}$ ) in a 5 L of container for 24 h, and then the carbon was taken out and packed into the HBTF, after mixed with nitrifying culture immobilized carbon.

### 2.2. Experimental setup

The studied HBTF (Fig. 1) has three segments, each with dimensions of  $15\text{ cm} \times 15\text{ cm}$  with 10 cm in length. The air stream containing  $\text{H}_2\text{S}$  and  $\text{NH}_3$  was directed from one side of the packing bed. The  $\text{H}_2\text{S}$  and  $\text{NH}_3$  inlet concentration was adjusted by mass flow controller (The Brooks Model 5850E, USA) at the outlet of a standard 5%  $\text{H}_2\text{S}$  and  $\text{NH}_3$  gas cylinder (Soxal Gas, Singapore) respectively, through mixing with air supplied by an air blower. Four sampling ports were located along the bed of the HBTF for gas sampling.

The recirculation solution was trickled down from the top of each segment, and the flow rate of the solution was controlled at  $1.2\text{ L min}^{-1}$ . The sump was maintained at 10 L and the recirculation solution was refreshed once every 5 days. The solution contained ( $\text{g L}^{-1}$ ):  $\text{KH}_2\text{PO}_4$ , 4.08;  $\text{K}_2\text{HPO}_4$ , 10.44;  $\text{NaHCO}_3$ , 2.00;  $\text{MgCl}_2\cdot 6\text{H}_2\text{O}$ , 0.46. The pH of the recirculation solution was controlled at 6.0–8.0

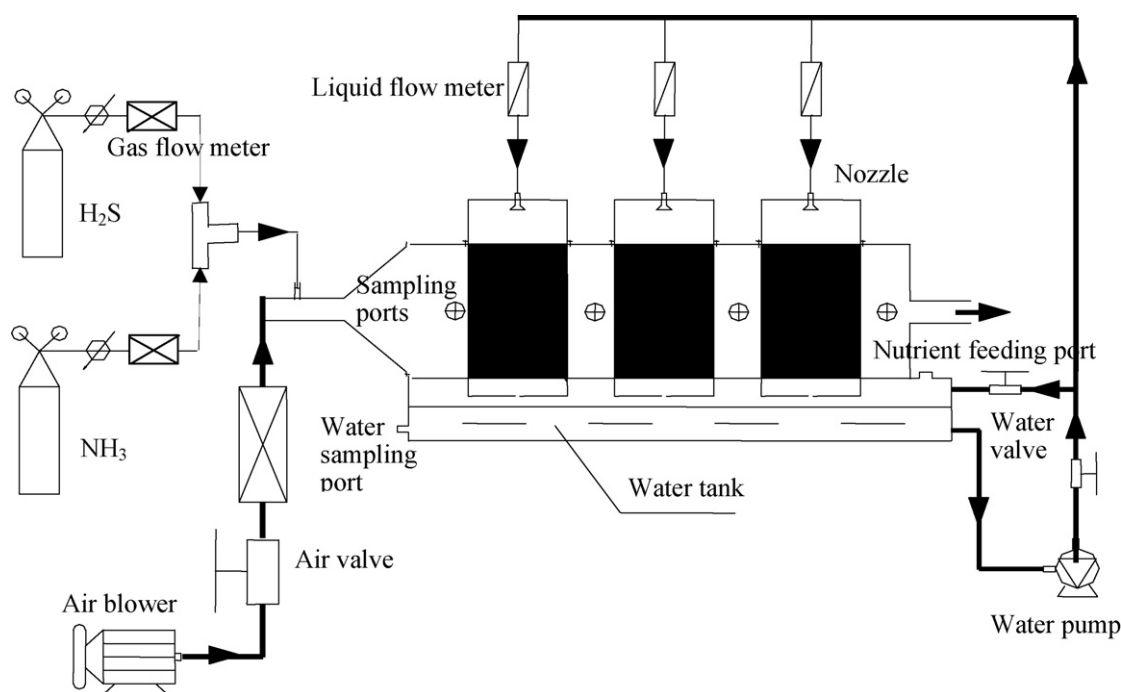


Fig. 1. Schematic diagram of the horizontal biotrickling filter (HBTF) system.

**Table 1**  
Experimental designs in the HBTF.

Phase	Period (d)	Inlet loading ( $\text{g m}^{-3} \text{h}^{-1}$ )	
		H <sub>2</sub> S	NH <sub>3</sub>
1	0–30	2–18	1–9
2	31–111	4–137	2–68
3	112–123	0	0
4	124–196	27–137	14–137
5	197–244	0	0
6	245–316	55	27

by adding 0.5N NaHCO<sub>3</sub> with an automatic pH controller (ON/OFF type, SIKO, PR75-C, Singapore). The system was operated at room temperature (about 25 °C) throughout the experiments. The bed pressures were measured using a water manometer with a minimum reading of 1-mm water column. The operating conditions are shown in Table 1.

### 2.3. Protein analysis

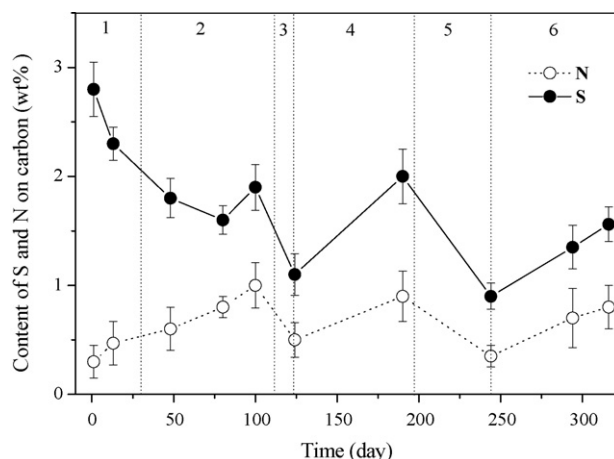
The amount of biomass on carbon was quantified based on the measurement of protein. Protein was extracted from microorganisms according to the methods of Kim and Deshusses [18]. About 1 g carbon sample was collected from bioreactors and placed in 5 mL of 1N NaOH solution in a tube, and the tube was pounded for 3 min. The solution with the carbon was then kept in a boiling water bath at 100 °C for 5 min to further extract biomass. The protein content in the solution was determined using the Bradford Protein Assay (Sigma, USA). The “clean” carbon (i.e. without biomass) was dried at 105 °C for 24 h to be ready for weighing. Biomass was reported as mg protein/g carbon.

### 2.4. SOUR

Biological activities of biofilm were described by determining oxygen uptake rate (OUR) [19]. About 5 mL of carbon sample taken from the HBTF was carefully washed with distilled water, and was put in a pre-cleaned BOD bottle. The BOD bottle was then fully filled with the pre-aerated mineral nutrient, and the oxygen-sensing probe with stirring mechanism (YSI 5000, Yellow Springs, OH) was immediately inserted into the BOD bottle. Endogenous respiration was first monitored. The substrate-induced OUR (SOUR) was determined after the addition of a concentrated solution to reach a final concentration of 50 mg L<sup>-1</sup> of S<sub>2</sub>O<sub>3</sub>-S and NH<sub>4</sub>-N for (SOUR)<sub>S</sub> and (SOUR)<sub>N</sub>, respectively. SOUR values were corrected for the endogenous respiration, and rates were normalized to the protein content of each sample.

### 2.5. Analytical methods

Surface pH of carbon was determined following the methods described previously [20]. The surface structures of carbon samples were determined using the Micrometrics BET Analyzer model ASAP 2010. The sulfur and nitrogen of carbon samples were determined using a thermo-analytical analyzer (PE2400 series II CHNS/O analyzer, PerkinElmer Instruments). Three analyses for each sample were carried out. Outlet H<sub>2</sub>S concentration was measured using a Jerome 631-X H<sub>2</sub>S analyzer (Arizona Instruments, USA) and inlet H<sub>2</sub>S concentration was determined using a Finch-con II portable H<sub>2</sub>S monitor (Infifitron, Seoul, Korea). Outlet NH<sub>3</sub> concentration was measured using a gas-detection tube (Draeger Safety Asia, Singapore). Inlet NH<sub>3</sub> concentration was determined using a BW NH<sub>3</sub> gas detector (model: GAXT-A, sensor: electrochemical cell, Calgary, AB, Canada).



**Fig. 2.** Content profiles of sulfur and nitrogen on carbon samples from the middle section of the HBTF.

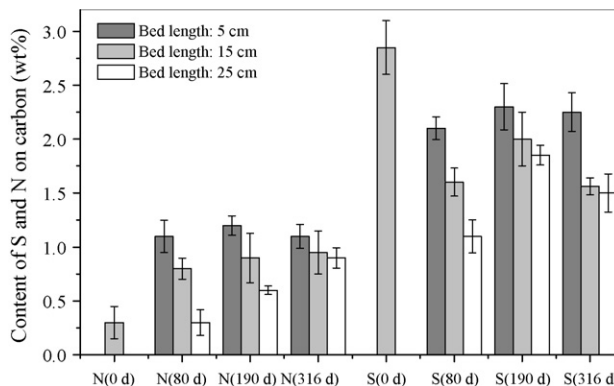
## 3. Results and discussion

### 3.1. Distribution of degradation products

The total sulfur and nitrogen removed by the HBTF system were 1508 and 899 g, while the total sulfur and nitrogen fed were 1670 and 925 g over 316 days of operation, respectively. The cumulative removal efficiencies were 90.3% and 97.1% for H<sub>2</sub>S and NH<sub>3</sub> respectively. The fate of the removed sulfur and nitrogen was evaluated through their distribution on carbon and in recirculation liquid.

The content profiles of sulfur and nitrogen on carbon samples in the HBTF are shown in Fig. 2. During the initial 80 days, the sulfur content decreased gradually, from 2.8 to 1.6 wt%. Meanwhile, an increasing trend was observed for the nitrogen element. It indicates that sulfur compounds pre-adsorbed on exhausted carbon were bio-regenerated, while some of NH<sub>3</sub> gas was adsorbed into the carbon. The contents of sulfur and nitrogen were relatively low after phases 3 and 5 without the supply of H<sub>2</sub>S and NH<sub>3</sub> (Fig. 2). The sulfur and nitrogen was accumulated at phase 4 with high loadings of H<sub>2</sub>S and NH<sub>3</sub>. Compared with that on exhausted carbon (7.8 wt% S), the contents of sulfur on carbon in the HBTF were much lower throughout the entire operation. The service life of exhausted carbon was extended greatly.

Fig. 3 shows the contents of sulfur and nitrogen on carbon along the HBTF bed. On day 80, the content of sulfur and nitrogen on carbon significantly decreased from the inlet (2.3 wt% S and 1.1 wt% N) to the outlet section of the HBTF (1.1 wt% S and 0.3 wt% N). The distribution could be attributed to the high concentration



**Fig. 3.** Content profiles of sulfur and nitrogen on carbon samples along the HBTF on days 80, 190 and 316.

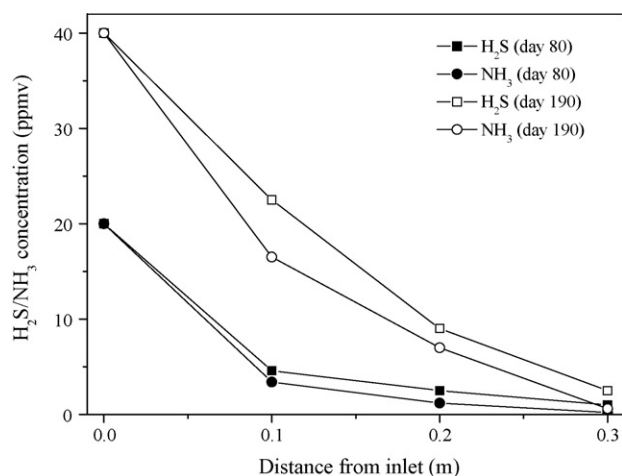


Fig. 4. Concentration profiles of H<sub>2</sub>S and NH<sub>3</sub> along the HBTF on days 80 and 190.

of pollutants in the inlet section and very low concentration of pollutants in the outlet section of the HBTF (Fig. 4). Most of pollutants were eliminated in the inlet section on day 80. Considering exhausted carbon as packing material in the HBTF, the phenomena suggests that high H<sub>2</sub>S concentration in the inlet section may provide little concentration gradient to drive desorption of pre-adsorbed sulfur from the carbon. In contrast, low concentration in the outlet may cause a large driving force for the release of sulfur compounds from the carbon. On days 190 and 316, the difference in the distribution of sulfur and nitrogen along the HBTF bed decreased. It was most likely attributed to the change of pollutants concentration profiles along the HBTF over extended operation (Fig. 4).

The content of degradation products on carbon (0.9–2.8 wt% S and 0.3–1.0 wt% N) was very low (Fig. 2). It suggests that removed pollutants were mostly washed into the recirculation liquid. In the recirculation liquid, a near stoichiometric production of sulfate, i.e. >95% in most cases, was achieved for H<sub>2</sub>S biodegradation in the HBTF. Meanwhile, input NH<sub>3</sub> was biodegraded into nitrite/nitrate with high ratio (>85% in most cases) in the HBTF system. H<sub>2</sub>S and NH<sub>3</sub> gases were removed mainly via the biodegradation in the system, rather than the physical/chemisorptions. In addition, input NH<sub>3</sub> can also be served as the nitrogen source for cell synthesis because the external nitrogen nutrient was not supplied in the system.

The low content of the degradation products on carbon bed benefited the stable operation of the HBTF. This is due to preventing the accumulation of degradation products on the packing in a toxic concentration, while the toxic accumulation was observed in biofilters previously [3,7,8]. The low accumulation of degradation products in the HBTF was mostly attributed to both the liquid trickling and almost complete biodegradation of H<sub>2</sub>S. Under these conditions, sulfate as the main product of H<sub>2</sub>S oxidation can be washed off from the bed more easily than elemental sulfur. In addition, frequently refreshing liquid medium was essential to discharge the biodegradation products out of the system.

It was reported that in a biofilter, the accumulated contents of sulfur and nitrogen on the packing increased to 44 and 5.5 wt% over 230 days of operation, respectively [3]. The accumulation of elemental sulfur and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> finally deactivated the whole system due to the diminution of active site and augmentation of the packing materials. In biofilters for treating NH<sub>3</sub> only, the accumulation of nitrogen products on the packing inhibited the NH<sub>3</sub> biodegradation process [7]. The accumulation even caused the removal of NH<sub>3</sub> to completely stop after 58-day operation [8].

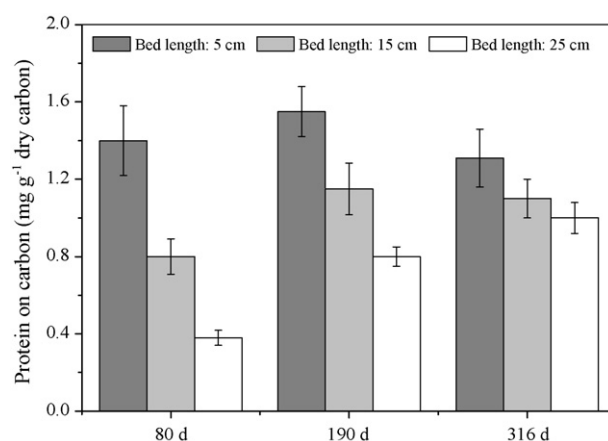


Fig. 5. Protein concentration of biofilm along the HBTF on days 80, 190 and 316.

### 3.2. Biomass accumulation

As shown in Fig. 5, the concentrations of biomass on carbon samples from the inlet section were ranged between 1.3 and 1.6 mg protein g<sup>-1</sup> dry carbon. They were higher than those from the outlet of the HBTF, i.e. 0.4–1.0 mg protein g<sup>-1</sup> dry carbon. The difference in biomass concentration along the bed decreased with operation time. This indicates that the biomass was distributed more even along the HBTF bed over extended operation. This was probably caused by high loadings of pollutants applied and the change of pollutants concentration profiles (Fig. 4).

The even distribution of biomass along the HBTF benefited the stable performance over a long-term operation. It was due to avoiding the problems of bed clogging and rapid activity loss caused by excessive biomass accumulated in the inlet section of bioreactors [11]. The biomass along the HBTF bed was distributed more even, compared with the vertical biofilter [11]. In a vertical VOC-degrading biofilter, the biomass concentration in the inlet section was 30 times higher than that in the outlet section [11]. However, in a vertical biotrickling filter for treatment of H<sub>2</sub>S, the difference in biomass concentration along the bed was insignificant [21]. The relatively even distribution of biomass along the HBTF bed could be partially attributed to continuously trickling liquid and the design of horizontal reactor.

Note that the issue of biomass overgrowth is typical of VOCs removal in bioreactors [11,22]. The clogging problems are usually not subject to biotrickling filters degrading H<sub>2</sub>S or NH<sub>3</sub> because of the slow growth rate of autotrophic microorganisms [1]. In addition, the period without the supply of H<sub>2</sub>S and NH<sub>3</sub> (phases 3 and 5) could also be beneficial to controlling the biomass accumulation [22].

However, over a long-term operation, it was still observed that the performance deteriorated due to the biomass accumulation in the HBTF bed on days 100, 190, 264 and 294 (data not shown). Carbon in each section in the HBTF was washed by 5 L of tap water to remove loosely attached biomass. The actions resulted in an immediate improvement in the removal of H<sub>2</sub>S and NH<sub>3</sub>. On day 264, H<sub>2</sub>S and NH<sub>3</sub> inlet loading and other operating conditions were maintained constant to allow a direct comparison. The removal efficiency of H<sub>2</sub>S was significantly increased from 77% to 90% and that of NH<sub>3</sub> was increased from 91% to 97% after removing this biomass. In a biomass balance analysis, nearly 1/3 of the total biomass in the bed was removed after washing carbon on day 264. Biomass accumulation could decrease specific surface area of packing bed because biomass may grow in the void fraction between the packing materials [23]. As a result, the available area for gas pollutants to diffuse into the biofilm decreased, which inhibited the

**Table 2**  
pH and surface structure of carbon samples in the HBTF.

Sample	Distance from the inlet (cm)	pH	Surface structure			
			$V_{mic}$ ( $\text{cm}^3 \text{g}^{-1}$ )	$S_{BET}$ ( $\text{m}^2 \text{g}^{-1}$ )	$S_{mic}$ ( $\text{m}^2 \text{g}^{-1}$ )	$S_{ext}$ ( $\text{m}^2 \text{g}^{-1}$ )
Day 80	5	6.7	0.153	705	355	340
	15	7.1	0.156	717	362	355
	25	7.4	0.171	727	365	362
	Average	7.1	0.160	716	361	352
Day 190	5	6.5	0.154	690	346	336
	15	6.9	0.163	697	353	345
	25	6.8	0.169	692	358	349
	Average	6.7	0.162	693	352	343
Day 316	5	6.6	0.160	680	356	324
	15	6.7	0.176	703	360	343
	25	6.4	0.163	684	344	340
	Average	6.5	0.166	689	353	336
FC		9.9	0.180	910	400	510
EC		6.1	0.133	772	306	455

$V_{mic}$ : micropore volume;  $S_{BET}$ : BET surface area;  $S_{mic}$ : micropore area;  $S_{ext}$ : external surface area; FC: fresh carbon; and EC: exhausted carbon.

biodegradation capacity [11,24]. Thus, it is still necessary to periodically remove some biomass from the filter to maintain a good performance over a long-term operation.

### 3.3. Biological activity of biofilm

Biological activity of biofilm in the HBTF was estimated by the substrate-induced oxygen uptake rate (SOUR), as shown in Fig. 6. It can be observed that the activities of biofilm on day 190 were lower than those on day 80. This may suggest that part of biomass in the biofilm in the HBTF was inactivated over extended operation.

On day 80, biological activity of sulfide-oxidizing biofilm (S) was the highest at the inlet section and decreased along the length of the HBTF (Fig. 6). In contrast, the activity of nitrifying biofilm (N) was the highest at the middle section of the HBTF on day 80. The phenomena could be explained by the inhibition of free ammonia due to high  $\text{NH}_3$  loadings at the inlet section and by the ammonium limitation at the outlet of bioreactors (Fig. 4). In addition, the low activity of nitrifying biofilm in the inlet may also be caused by high loading of  $\text{H}_2\text{S}$  (Fig. 4) and high sulfur content in the section (Fig. 3). On the other hand, although the activity of nitrifying biofilm in the inlet section was influenced negatively, the removal of  $\text{NH}_3$  gas was not affected (Fig. 4). The removal of  $\text{NH}_3$  gas might be partially attributed to the absorption/adsorption or chemical reaction in the inlet section.

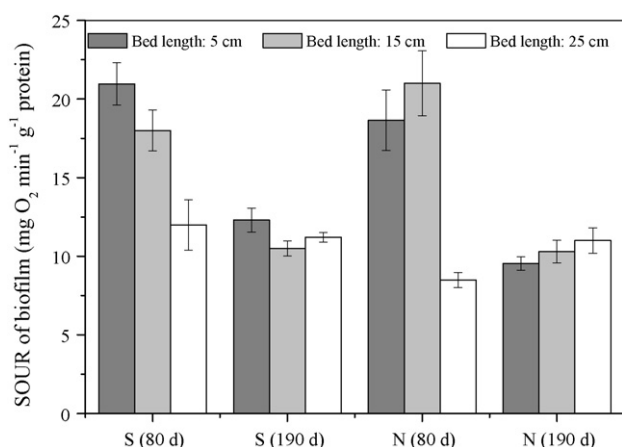


Fig. 6. Biological activity of biofilm along the HBTF on days 80 and 190.

On day 190, biological activity of biofilm along the HBTF became more even than that on day 80 (Fig. 6). The spatial change was possibly due to high loadings of pollutants applied during this period and the change of pollutant concentration profiles along the HBTF (Fig. 4). In addition, the change indicates that certain nitrifying culture might be acclimatized by high concentration of  $\text{NH}_3$  and  $\text{H}_2\text{S}$  in the inlet section.

### 3.4. Change of carbon surface characteristics

The pH values and surface structures of carbon samples from the HBTF were identified (Table 2). It can be seen that the pH values of carbon samples in the HBTF were near neutral. This overcomes the problem of the system acidification which usually happened in biofilters [5,25]. The neutral pH values are better for the adsorption of  $\text{H}_2\text{S}$  gas firstly and then for further biodegradation in biofilters [25], even though  $\text{H}_2\text{S}$  can also be effectively biodegraded under low pH as reported by some studies [9,15]. Moreover, the neutral pH range was proper for the growth of nitrifying biofilm. In addition, the pH values of carbon from each section along the HBTF bed were almost identical (Table 2). This is possibly attributed to the design of horizontal reactor and liquid trickling.

For surface structure of carbon samples, micropore volume ( $V_{mic}$ ) and surface area ( $S_{mic}$ ) remained at similar level while external surface area ( $S_{ext}$ ) slightly decreased over 316 days of operation (Table 2). With regards to the distribution along the HBTF bed, the  $S_{ext}$ ,  $V_{mic}$  and  $S_{mic}$  increased slightly from the inlet to outlet section of the bed on day 80. Over extended operation, surface structures of carbon samples along the HBTF bed became even. This result was consistent with the distribution of biodegradation products (Fig. 3) and of biomass (Fig. 5) on carbon along the HBTF.

The changes of carbon surface characteristics will determine the lifetime of exhausted carbon in this study. It was observed that the  $V_{mic}$  and  $S_{mic}$  of carbon in the HBTF remained at relatively stable (Table 2). This indicates that the biodegradation played a major role in the removal of pollutants while the adsorption was only an intermediate process. The adsorption step is essential; otherwise, the biodegradation cannot happen at all. In addition, the available micropores of carbon let the adsorption still happening during shock loadings. This significantly contributed to the high performance in the HBTF over a long-term operation. Compared with those of exhausted carbon ( $0.133 \text{ cm}^3 \text{ g}^{-1}$  and  $306 \text{ m}^2 \text{ g}^{-1}$ ), the  $V_{mic}$  and  $S_{mic}$  of carbon samples in the HBTF ( $0.153\text{--}0.176 \text{ cm}^3 \text{ g}^{-1}$

and 344–365 m<sup>2</sup> g<sup>-1</sup>) were higher. These carbons were still far from exhausted again until 316 days of operation.

### 3.5. Pressure drop

The pressure drop in the HBTF was ranged between 3 and 12 mm H<sub>2</sub>O m<sup>-1</sup> bed over 316 days of operation. The pressure drop decreased evidently after carbon was water washed on days 100, 190, 264 and 294 (data not shown). This indicates that biomass accumulation has a negative effect on pressure drop. After 280 days of operation, the superficial velocity was changed gradually from 0.005 to 0.14 m s<sup>-1</sup>, corresponding to EBRT of 60–2 s. There was a good linear relationship between the pressure drop and superficial velocity (data not shown). Moreover, the pressure drop was only 16 mm H<sub>2</sub>O m<sup>-1</sup> even at a high superficial air flow rate of 0.14 m s<sup>-1</sup> (2 s of EBRT).

The pressure drop in the HBTF was lower, compared with other biofilters, e.g., 20–88 mm H<sub>2</sub>O m<sup>-1</sup> [26], 45–93 mm H<sub>2</sub>O m<sup>-1</sup> [27], up to 204 mm H<sub>2</sub>O m<sup>-1</sup> [4]. A high pressure drop will result in high energy consumption requirements to maintain the good performance of a bioreactor. Pressure drop is dependent on the geometry of the packing materials, reactor configuration, biodegradation products and biomass accumulation, etc. The low pressure drop in this study could be the result of a combination of several respects. Firstly, the horizontal design makes the pressure drop low even though the bulk density of activated carbon was high [28]. Secondly, compared with heterotrophs, a strictly autotrophic is preferred to avoid clogging problems [9]. Thirdly, H<sub>2</sub>S was almost completely bio-oxidized into sulfate, which can be washed from the system easily. Moreover, activated carbon had good mechanical strength, which led to negligible bed compression and short-circuiting during operating periods [26].

## 4. Conclusions

The results show that the contents of biodegradation products on carbon were low, i.e. 0.9–2.8 wt% S and 0.3–1.0 wt% N. This benefits the stable operation of the HBTF, due to preventing the accumulation of metabolic products in a toxic concentration. The biomass was distributed evenly along the bed. This avoids the problem of bed clogging and activity loss. The pH values of carbon in the HBTF were controlled at neutral, which benefited simultaneous biodegradation of H<sub>2</sub>S and NH<sub>3</sub>. In addition, micropore structure of carbon remained relatively stable, and the available micropores of carbon let the adsorption still happening during shock loadings. The pressure drop in the HBTF was very low. These advantages of the HBTF system significantly contributed to the high performance in the HBTF over a long-term operation.

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